國立臺灣大學工學院環境工程學研究所

碩士論文

Graduate Institute of Environmental Engineering

National Taiwan University

Master Thesis

飲用水配水管線中鉛和不鏽鋼之電化腐蝕反應

Galvanic Corrosion between Lead and Stainless Steel in
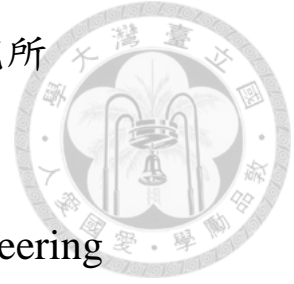
Drinking Water Distribution Systems

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中華民國 105 年 1 月

January, 2016

摘要

臺灣自 1979 年開始禁止使用鉛管作為飲用水管,而為了改善飲用水的品質,自來水公司開始使用進行管線的汰換工程,不鏽鋼管為近十年選用的新管材。由於考慮到汰換管線的成本和住家的隱私問題,因此汰換工程主要選擇在公共給水管線中進行,家用鉛管則繼續殘留在住戶中,而造成部分汰換的狀況產生。由於鉛和不鏽鋼之間具有標準電位差,因此當鉛和不鏽鋼連接送水時會產生電化腐蝕反應。

此研究的目的是要了解在不同的操作條件下,如:pH 值、[氯離子濃度 mg/L]/[硫酸根離子濃度 mg/L] 或 CSMR 和在相同 CSMR=2 時改變氯離子和硫酸根離子濃度,鉛和不鏽鋼之間的電化腐蝕反應對於鉛溶出的影響,此外評估使用正磷酸作為腐蝕抑制劑控制鉛溶出的效果。實驗結果顯示,當鉛和不鏽鋼的電化腐蝕反應產生時,水中的鉛濃度會提高,且當 pH 降低或 CSMR 提高時,鉛溶出的影狀況會更明顯。此外,在自水中加入正磷酸鹽可以減緩鉛溶出的情況,但是當電化腐蝕反應發生時,仍需注意 CSMR,因為即使在飲用水中含有正磷酸鹽的情況下,CSMR 越高飲用水中的鉛濃度仍然越高。
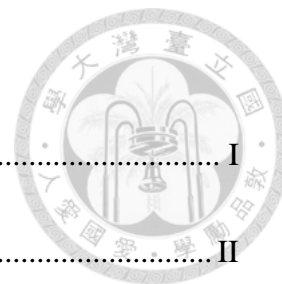

關鍵字: 鉛,不鏽鋼,電化腐蝕,pH,CSMR,正磷酸鹽

Abstract

Lead pipe has been banned for distributing drinking water in Taiwan since 1979. In order to improve drinking water quality, partial replacement of lead pipes, in which lead pipes in the public area were replaced by stainless steel pipes while those in the private premises remained intact, was commonly practiced in Taiwan due to the high cost and difficulties to access private properties. Due to the different potentials of lead and stainless steel, galvanic corrosion may take place if they are electronically connected in water. The objectives of this research are to characterize the effects of galvanic connection between lead and stainless steel on lead release and the effects of pH, chloride and sulfate concentration on this process. The experiments were conducted by connecting aged lead pipes and stainless steel fittings under different pH, chloride to sulfate mass ratio or CSMR and different chloride and sulfate concentration under a fixed CSMR of 2. Orthophosphate was evaluated as a corrosion inhibitor to control lead release. The results demonstrated that lead release increased when lead and stainless steel were galvanically connected and the rate of lead release accelerated with the decreasing pH and increasing CSMR. Orthophosphate could effectively reduce lead release but CSMR needs to be considered since water with a higher CSMR still caused more lead release when galvanic corrosion took place.

Keywords: Lead, Stainless steel, Galvanic corrosion, pH, CSMR, Orthophosphate

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Chapter 1 Introduction

1.1   Background

Lead is a toxic metal. Long term exposure of lead can cause physical and mental development problems for children and high blood pressure, kidney failure and reproductive problems for adults (WHO, 1993). Although lead pipe has been banned in many countries in the 1980s, it is still used in plumbing materials such as brass fittings and faucets because of its ductility and ability to enhance the machinability of alloys. Lead in drinking water is regulated by the USEPA with an action level of 15 μg/L (Lead and Copper Rule, 1991) while the WHO guideline value is 10 μg/L. The standard of lead in Taiwan was revised from 50 μg/L to 10 μg/L in 2013 (Taiwan EPA, 2008). The use of lead pipe in the drinking water distribution system was banned in Taiwan in 1979. In 2002, the Taipei Water Department announced that 625 km of lead pipes still remained in Taipei and commenced to replace lead pipe with stainless steel pipe in the public areas. Those in the premises were not replaced due to the high cost and difficulties to access private properties. According to the Taipei Water Department, 2.7 % to 6 % of the lead service lines were replaced annually (Chinese Taiwan Water Work Association, 2007) and by 2015 around 157 km lead pipes still existed in the distribution system. Under such partial replacement circumstances, galvanic corrosion

between lead and stainless steel might occur. It has been reported that significant lead

release may result from galvanic corrosion between lead and copper and endanger

public health (Frumkin, 2010; Brown, 2011).
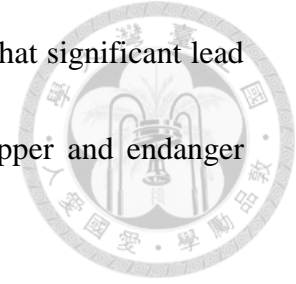
1.2   Research objectives

It is hypothesized that galvanic corrosion between lead and stainless steel can

cause significant amount of lead released into drinking water. The objectives of this

thesis are:
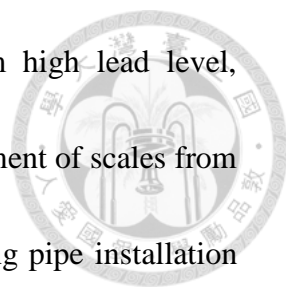
1. To investigate the effects of solution pH, chloride and sulfate concentrations on

    lead release due to galvanic corrosion between lead and stainless steel pipes.

2. To evaluate the effectiveness of using orthophosphate as a corrosion inhibitor to

    control lead release due to galvanic corrosion between lead and stainless steel.

Chapter 2 Literature review

2.1 Partial replacement of lead service line

Lead pipes were used in Taiwan before they were banned in 1979. Although most of the lead pipes in the public water mains had been replaced, there are still lead service lines (< 20 m) remaining between the street pipe and water meter and inside the households (after water meter). The section of service line that extends to the building belongs to private property which public utility cannot access and replace. The additional cost to replace lead pipes in private property also discourages consumers from doing so; hence many buildings still contain sections of lead pipes after partial replacement. It has been reported that partial replacement of lead pipes, unexpectedly, may increase lead concentration in drinking water. Zietz et al. showed that 50% of households in Germany with elevated lead concentration were related to partial lead service line replacement (Zietz et al., 2001). In 2010, the Centers of Disease Control and Prevention (CDC) in the US warned that children were more likely to have high blood lead level (greater than or equal to 10 μg/dL) when lead service lines are partially replaced as compared to those who live in buildings with either service lines without lead or undisturbed lead service lines (Frumkin, 2010). Lead contamination due to partial replacement can be short-term that lasts for days to

weeks and long-term that lasts for months to years. Short-term high lead level, particularly resulting from particulate lead, is caused by the detachment of scales from the inner pipe surfaces in the premise side that are disturbed during pipe installation (Schock et al. 1996). In a laboratory study employing continuous flow, Boyd et al. (2004) scarped lead pipes at one end to simulate the disturbance during partial replacement and found that the initial total lead concentration was greater than 1000 µg/L, then gradually reduced to below the action level after 30 min of flushing. The time required to lower the lead level to the acceptable level increased with decreasing flow rate, which may take up to 144 hr. Sandvig et al. (2008) showed that the total lead levels in the first-liter draw samples were reduced in two months after full replacement of lead source but the lead concentration remained high in those of partial replacement. Long-term lead contamination can be caused by galvanic corrosion of lead due to the connection between old lead pipe and the newly installed dissimilar metallic pipe or fitting. It has been reported that lead release from the partial replaced rigs was greater than that from pure lead service line, no matter how many percent of the lead pipes were replaced by copper (Triantafyllidou and Edwards, 2010). Wang et al. (2012) also reported that after 6hr stagnation, 4-6 times higher of total lead were found in the systems that aged lead pipes were galvanically connected to copper pipes.

2.2 Galvanic corrosion

Galvanic corrosion is an electrochemical process that occurs when two or more different conducting materials are electrically connected to each other in an electrolyte (Burkert, 2002). Corrosion (oxidation) takes place at the anode which is related to the galvanic current by Faraday's law while the cathode is protected (reduction) (Zhang, 2011). In the US, Canada and UK, partial replacement is commonly practiced and lead pipe has been replaced by copper pipe (USEPA, 1986; Health Canada, 2009; DWI, 2010). When lead is connected to copper, lead acts as the anode and copper acts as the cathode (Figure 1). Oxidation at the lead anode produces Pb^{2+}, a Lewis acid, which causes a pH drop at the lead surface due to the formation of soluble hydroxide complexes ($Pb^{2+} + H_2O \rightarrow PbOH^+ + H^+$) (Stumm and Morgan, 1996) or insoluble precipitates that contain OH^- (Dudi, 2004). This pH drop at the lead surface could generate a local low pH zone, which further accelerates the rate of galvanic corrosion. On the contrary, reduction at the copper cathode produces hydroxide ions which increase the solution pH and protects the copper pipe from corrosion. (Nguyen et al., 2010).
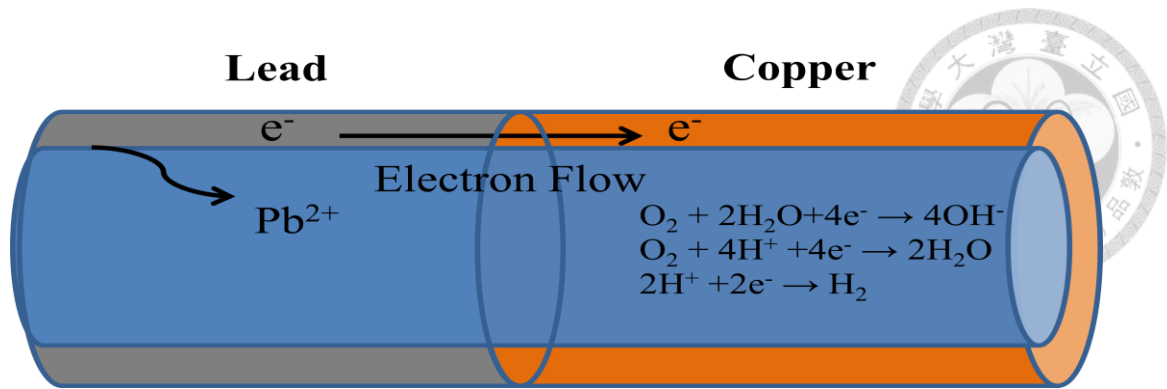
Figure 1 Galvanic connection between lead and copper.

Galvanic corrosion can be affected by several factors such as electrolyte properties, material's surface conditions, metallurgical factors and standard electrode potential between the metals (Zhang, 2011). The galvanic series ranking metals in the order of relative nobility in drinking water and seawater are shown in Table 1 and 2, respectively (Matsukawa et al., 2011; Fontana and Greene, 1978). The galvanic series was developed based on standard electrode potential, which could vary in different solutions. In a two-metal connection, the smaller of the difference between the standard potentials is, the weaker the galvanic effect is and vice versa (Perez, 2004). In the galvanic series, the metal that is less noble has a higher tendency to corrode. The relative nobility of lead and stainless steel does not change in seawater and drinking water.

Table 1 Galvanic series in drinking water (Matsukawa et al., 2011).

Noble or cathodic	
Stainless steel 304, Stainless steel 316	
Titanium, Copper, Bronze	↑ Cathodic
90-10 Cu Ni	
70-30 Cu Ni	
Brass	
Tin	
Lead	
Aluminum	
Carbon steel	
Gray cast iron	
Ductile cast iron	
Zinc	↓ Anodic
Magnesium	
Active or anodic	

Table 2 Galvanic series of some commercial metals and alloys in seawater (Fontana and Greene, 1978)
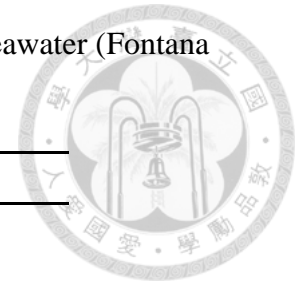
Noble or cathodic	
Platinum	
Gold	
Graphite	↑ Cathodic
Titanium	
Silver	
Chlorimet 3	
Hastelloy C	
18-8 Mo stainless steel (passive)	
18-8 stainless steel (passive)	
Chromium steel > 11% (passive)	
Inconel (passive)	
Nickel (passive)	
Silver solder	
Monel	
Bronzes	
Copper	
Brasses	
Chlorimet 2	
Hastelloy B	
Inconel (active)	
Nickel (active)	
Tin	
Lead	
Lead-tin solders	
18-8 Mo stainless steel (active)	
18-8 stainless steel (active)	
Ni-resist Chromium steel >11% Cr (active)	
Cast iron	
Steel or iron	↓ Anodic
2024 aluminum	
Cadmium Commercially pure aluminum	
Zinc	
Magnesium and its alloys	
Active or anodic	

2.3 Chloride to sulfate mass ratio (CSMR)
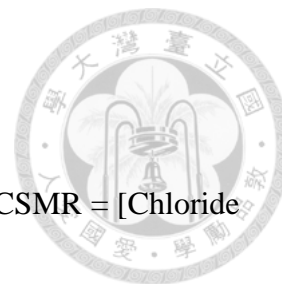
CSMR is the ratio of chloride and sulfate mass concentration (CSMR = [Chloride mass concentration] / [Sulfate mass concentration]). Oliphant first reported that chloride tends to promote lead release when lead is galvanically connected to copper (Oliphant, 1983). In 1985, Gregory introduced the CSMR concept and demonstrated the importance of CSMR on galvanic corrosion of lead, in which solution with a high CSMR would promote galvanic corrosion, whereas that with a low CSMR would reduce its rate (Gregory, 1985). In a utility survey, Edwards et al. (1999) showed that 64 percent of the utilities with a CSMR greater than 0.58 having lead levels exceeded the action level and suggested that utilities should control the CSMR less than 0.58 to prevent galvanic corrosion. Dudi reported that lead leaching from yellow brass and red brass increased with the increasing CSMR in 11 of 12 tests (Dudi, 2004). In a study conducted by Triantafyllidou and Edwards (2010), lead pipes were connected to copper pipes in order to determine the effects of galvanic corrosion on lead release. Their results showed that 3 to 12 times more lead released was observed under a high CSMR (CSMR=16) compared to the water with a low CSMR (CSMR=0.2). CSMR could be increased during water treatment such as switching coagulant from alum to ferric chloride and polyaluminium chloride (Edwards and Triantafyllidou, 2007), using a magnetic ion exchange (MIEX) resin to remove anions or NOM (Boyer et al.,

2008; Willson and Boyer, 2012), and blending treated water with desalinated water (Liu et al., 2010).

2.4 Orthophosphate as a corrosion inhibitor

Phosphate-based corrosion inhibitors including orthophosphate, zinc orthophosphate, polyphosphate have been used to prevent both iron and lead corrosion (Dodrill and Edwards, 1995). McNeill and Edwards (2002) surveyed the US drinking water utilities and found that utilities which use orthophosphate for corrosion control increased from 2% to 20%. Edwards and McNeill (2002) investigated the effects of orthophosphate on lead release using new lead pipe and aged pipes under different pH and alkalinity. Their results showed that orthophosphate reduced 70% of soluble lead concentration when compared to conditions without orthophosphate. Soluble lead can form precipitates with orthophosphate such as hydroxypyromorphite $(Pb_5(PO_4)_3OH)$, tertiary lead orthophosphate $(Pb_3(PO_4)_2)$ and chloropyromorphite $(Pb_5(PO_4)_3Cl)$. Lead phosphate minerals are less soluble than lead carbonate (Nriagu, 1974; Schock, 1989). Hence, the pipe is more stable when coated with these phosphate precipitates. The effectiveness of orthophosphate to control soluble lead release, however, depends on water chemistry. At very low dissolved inorganic carbon (DIC) concentration, the optimum pH for lead phosphate formation is around 8 and

the optimum pH decreases with the increasing DIC, with the lowest pH around 7 (Schock, 1989; Tam and Elefsiniotis, 2009).   Nguyen et al. (2011) showed that when sulfate concentration was less than 10 mg/L, the addition of orthophosphate might enhance lead release. In addition, orthophosphate can be effective in reducing soluble lead, but not total lead in drinking water (Xie and Giammar, 2011).
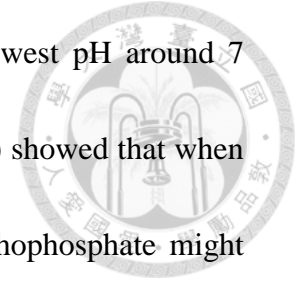
Chapter 3 Materials and Methods

3.1   Materials and chemicals

Aged lead pipes were harvested from the drinking water distribution system in Taipei City and supplied by the Taipei Water Department. The inner diameter of the lead pipe is 24 mm and the outer diameter is 33 mm. Harvested lead pipes were cut into 6 cm sections (Figure 2(a)). Locally purchased stainless steel 304 fitting (Figure 2(b)) was used in this study. In order to simulate the situation in the distribution system, the stainless steel fitting was directly connected to the harvested lead pipe to form a lead-stainless steel galvanic couple (Figure 2(c)). Stainless steel 304 has a composition shown in Table 3 (ASTM, 2004). Aged lead pipe sections were conditioned with synthesized tap water (Table 4) for 12 weeks before they were used to form the galvanic couple. The synthesized tap water was prepared using deionized water (ELGA, UK) and sodium chloride (Merck, Germany), magnesium sulfate heptahydrate (Merck, Germany), sodium bicarbonate (Merck Germany), calcium chloride dihydrate (Merck, Germany) and sodium hypochlorite (Sigma-Aldrich, USA).

The scales on the inner surfaces of the pipes before galvanic corrosion experiments were collected and analyzed using SEM (Nova NanoSEM 230, FEI,

USA) and XRD (Rigaku, Japan).
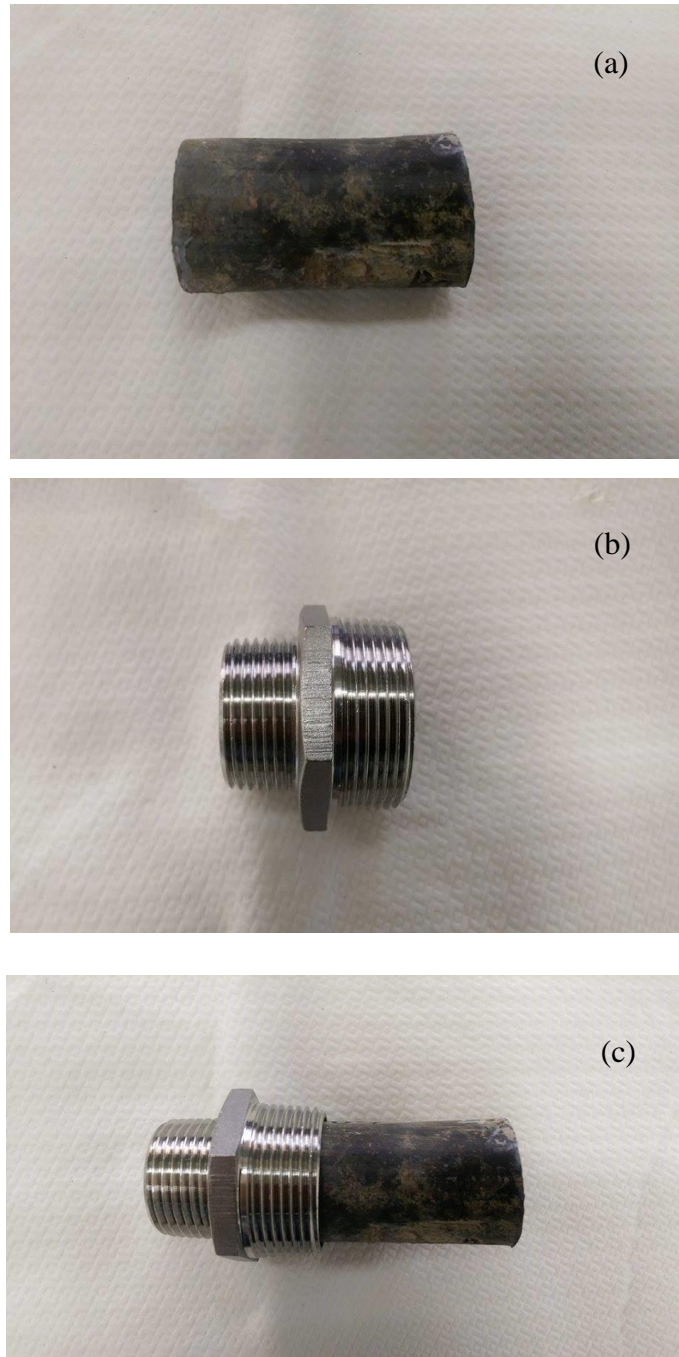
Figure 2 (a) 6 cm aged lead pipe section. (b) Commercially available stainless steel 304 fittings (c) Lead-stainless steel galvanic couple

Table 3 Standard composition of stainless steel 304

Element	C	Mn	P	S	Si	Cr	Ni	N	Fe
Maximum	0.08%	2%	0.045%	0.03%	0.75%	20%	10.5%	0.1%	Balance
minimum						18%	8%		Balance
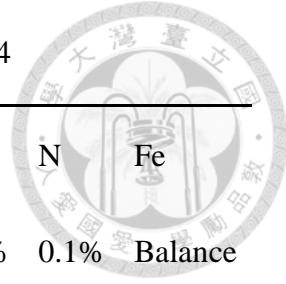
Table 4 Synthesized tap water composition

pH	7.45~7.55
Total Alkalinity	20 mg/L as $CaCO_3$
Total hardness	35 mg/L as $CaCO_3$
Chloride	15 mg/L
Sulfate	30 mg/L
Initial free chlorine	0.8 mg/L as Cl_2

3.2 Experimental setup

A dump-and-fill protocol was adopted in this study (Edwards and Triantafyllidou, 2007). Briefly, the galvanic couple was filled with the synthesized water, sealed with no headspace using parafilm and stagnant for 24 h on weekdays and 72 h over weekends. The water was then dumped and refilled with a fresh supply of synthetic water. The effects of pH, chloride and sulfate concentrations on lead release were determined by comparing results obtained from experiments under different conditions shown in Table 5. Solution pH was adjusted using 0.1 M nitric acid and 0.1 M NaOH. After 9 weeks of experiment, orthophosphate was introduced into selected conditions (conditions 2, 4, 5 and 6 in Table 5) to examine the effects of orthophosphate on lead release during the galvanic corrosion. The concentration of orthophosphate used in the experiment was fixed at 1 mg/L as P. Orthophosphate addition was ceased after 5 weeks and the experiment continued for another 4 weeks. Solution pH, soluble and total lead were measured. All experiments were conducted in duplicate.

Table 5 Test water conditions

	pH	[Cl$^-$] (mg/L)	[SO$_4^{2-}$] (mg/L)	CSMR	Conductivity (μs/cm)
Condition 1	6.5	15	30	0.5	169.5
Condition 2	7.5	15	30	0.5	167.3
Condition 3	8.5	15	30	0.5	168
Condition 4	7.5	6	30	0.2	165.1
Condition 5	7.5	240	30	8	905.8
Condition 6	7.5	60	30	2	421.3
Condition 7	7.5	120	60	2	585.4
Condition 8	7.5	250	125	2	1147

3.3 Analytical methods

Total lead and soluble lead were measured using inductively coupled plasma optical emission spectrometry (ICP-OES, Agilent 700 series) in accordance with Standard Method 3125-B. After the experiment, 5 mL sample was drawn from the completely mixed solution and filtered through a 0.2 μm pore size nylon filter. The filtrate was measured for soluble lead. The remaining sample was acid digested by adding 5 % v/v nitric and heated at 85°C for at least 2 h prior to total lead measurement. The solution pH was measured using a SUNTEX pH meter (sp2100) precalibrated using standard pH 4, 7, and 10 buffer solutions (Merck, Germany). Free chlorine concentration was determined following the DPD-FAS method in accordance to Standard Method 4500-Cl F. Scales scratched from the aged lead pipes were prepared for PbO_2 measurement based on a two-stage Iodometric method (Zhang et al., 2010).

Chapter 4 Results and discussions

4.1 Scale analysis of the aged lead pipe

An aged lead pipe was cut open as shown in Figure 3. Reddish scales were found on the pipe inner surfaces. The scales were scraped using a stainless steel spatula and prepared for SEM and XRD analyses.

The SEM image (Figure 4) showed that the scales were aggregates comprising crystals with different shapes and sizes, including plate-shape particles and cubic-shape nanoparticles, which were the characteristic shape and size for hydrocerussite ($Pb_3(CO_3)_2(OH)_2$) and scrutinyite (α-PbO_2)/plattnerite (β-PbO_2), respectively (Liu et al., 2008; Xie, 2010; Wang et al., 2012). The XRD pattern of the scales along with the standard patterns for α-PbO_2, β-PbO_2, $PbCO_3$ and $Pb_3(CO_3)_2(OH)_2$ are shown in Figure 5. Compared the XRD pattern of the scale with those of the standards, it can be concluded that β-PbO_2 is one of the dominant phases present in the scale. This observation was consistent with the fact that chlorine has been used for disinfection in Taipei and chlorine is able to oxidize Pb(II) species to form PbO_2 (Lytle and Schock, 2005). The mass concentration of PbO_2 in the scales was tested using the two-stage Iodometric method (Zhang et al., 2010) and the result showed that PbO_2 accounted for around 29% of the total lead in the scales. The rest of

the total lead could be $Pb_3(CO_3)_2(OH)_2$ and $PbCO_3$ as indicated by the plate-shape

crystal and the XRD pattern.

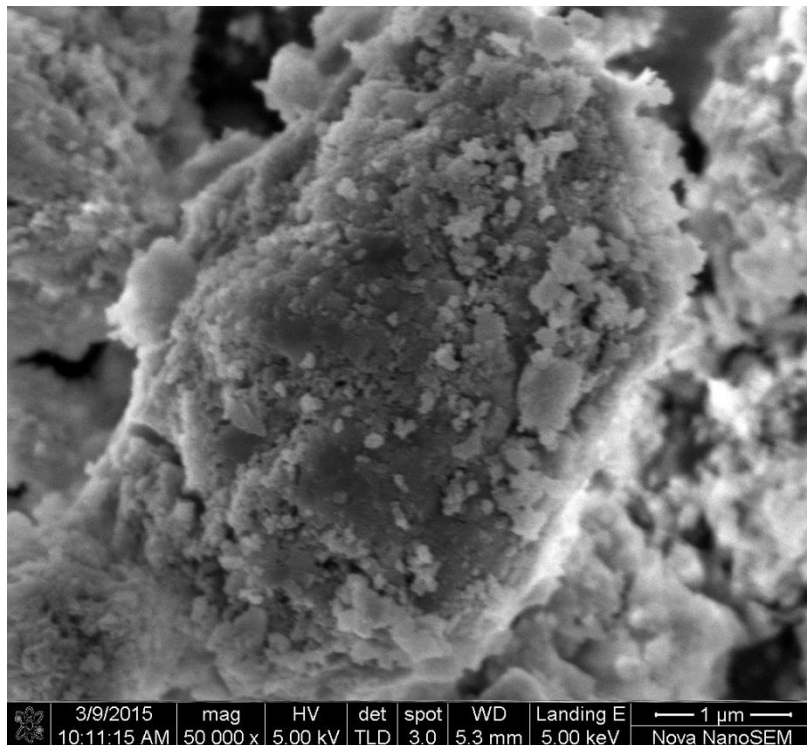Figure 3 Aged lead pipe which was cut horizontally before experiment
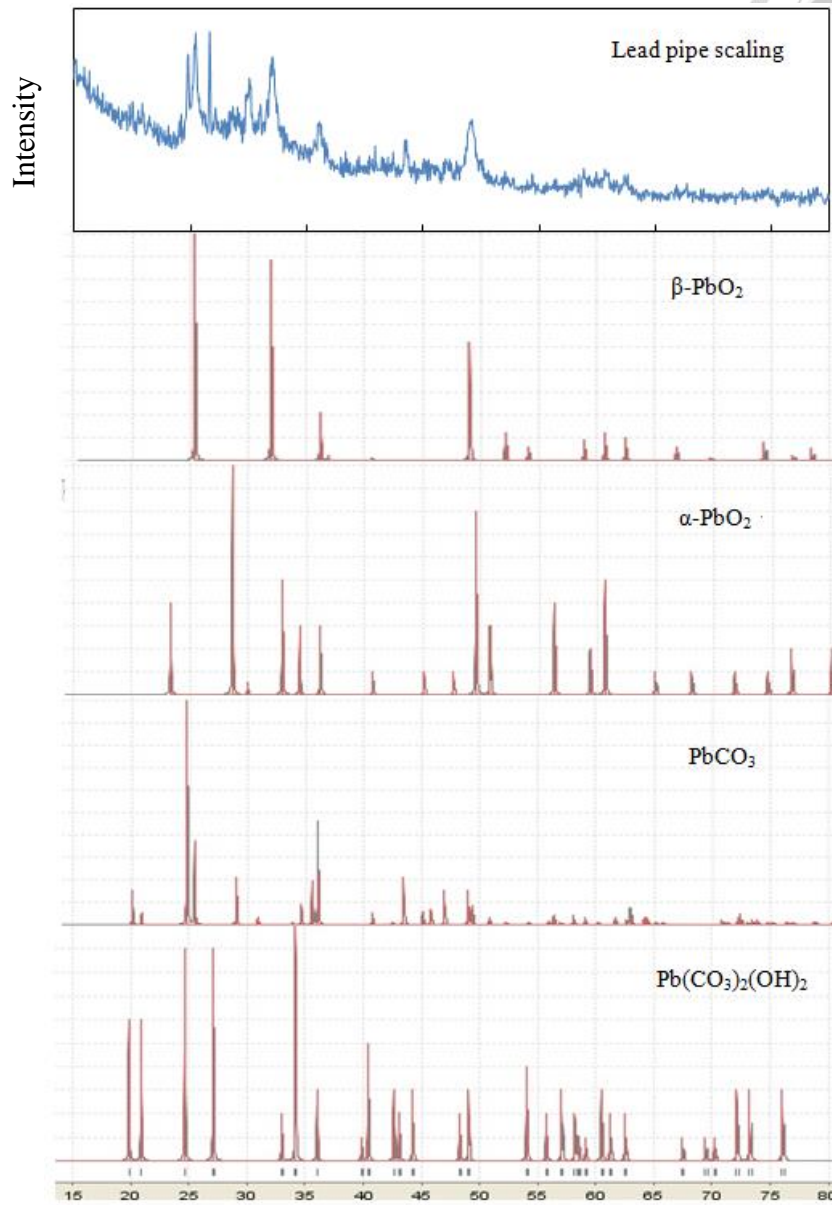

Figure 4 SEM images for scale collected from the aged lead pipe.

Figure 5 X-ray diffraction (XRD) pattern of the scale collected from aged lead pipe

and standard patterns for β-PbO$_2$, α-PbO$_2$, PbCO$_3$ and Pb$_3$(CO$_3$)$_2$(OH)$_2$.

4.2　Lead release due to galvanic corrosion

After 12 weeks of conditioning, aged lead pipes were connected to stainless steel fittings. Control experiments without galvanic connection were also conducted. Figure 6 shows the results of soluble and total lead concentrations for experiments conducted at pH=7.5 and CSMR=0.5. Both soluble lead and total lead were higher if aged lead pipes were connected to stainless steel fittings and the difference in total lead was much more significant than that in soluble lead. The Pb^{2+} released form the electrochemical reactions between the galvanic couple could form Pb(II) precipitates such as cerussite, hydrocerussite and lead sulfate if their solubility products were exceeded. Figure 7 shows the particulates collected from the galvanic experiments, in which white particles were observed. The precipitation could reduce soluble lead concentration and being detected as particulate fraction of the total lead, therefore, resulting in the greater difference observed in total lead. Soluble lead only contributed about 5% of total lead. In addition, lead release was positively correlated with the stagnation time, i.e., lead release data obtained after weekend (3-day stagnation) were generally greater than those obtained during the weekday (1-day stagnation). This observation could be related to the "dumped and filled" protocol used in this study
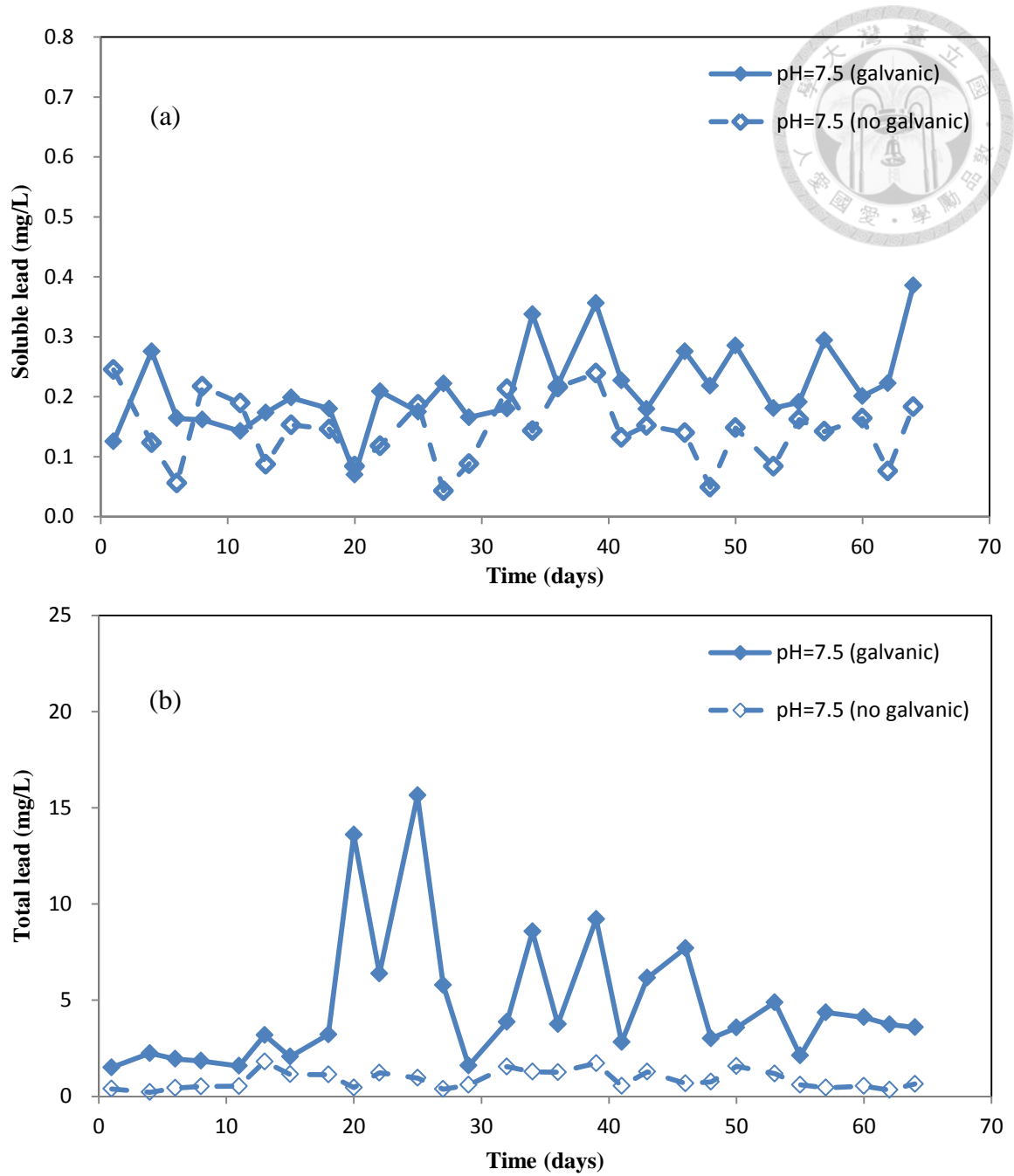
Figure 6 (a)Soluble lead concentration. (b)Total lead concentration measured with and without galvanic corrosion at pH=7.5 and CSMR=0.5.

Figure 7 White particles collected from the solution contacting with lead pipe-stainless steel fitting connection.

4.3 The effects of pH on galvanic corrosion.

Figure 8 shows the soluble lead concentration and total lead concentration for the samples collected from the experiments conducted at pH=6.5, 7.5 and 8.5 with a fixed CSMR=0.5. It was found that both soluble and total lead, in general, were higher when the experiments were conducted under a more acidic condition. The lead levels were higher when galvanic corrosion existed except a few soluble lead measurements. In the galvanic couple, the increase in proton concentration can accelerate the electrochemical reaction in the cathode ($O_2 + 4H^+ + 4e^- \rightarrow 2H_2O$, $2H^+ + 2e^- \rightarrow H_2$) (Figure 1) and enhance the overall lead release. The higher soluble lead released then promoted the precipitation of lead particles. It should be noted that, however, the predominant lead solid could be different at different pH values. For example, cerussite ($PbCO_3$) is more thermodynamically favorable to form at pH <7.5, while hydrocerussite ($Pb_3(CO_3)_2(OH)_2$) predominantes at pH greater than 7.5 (Lytle and Schock, 2005).
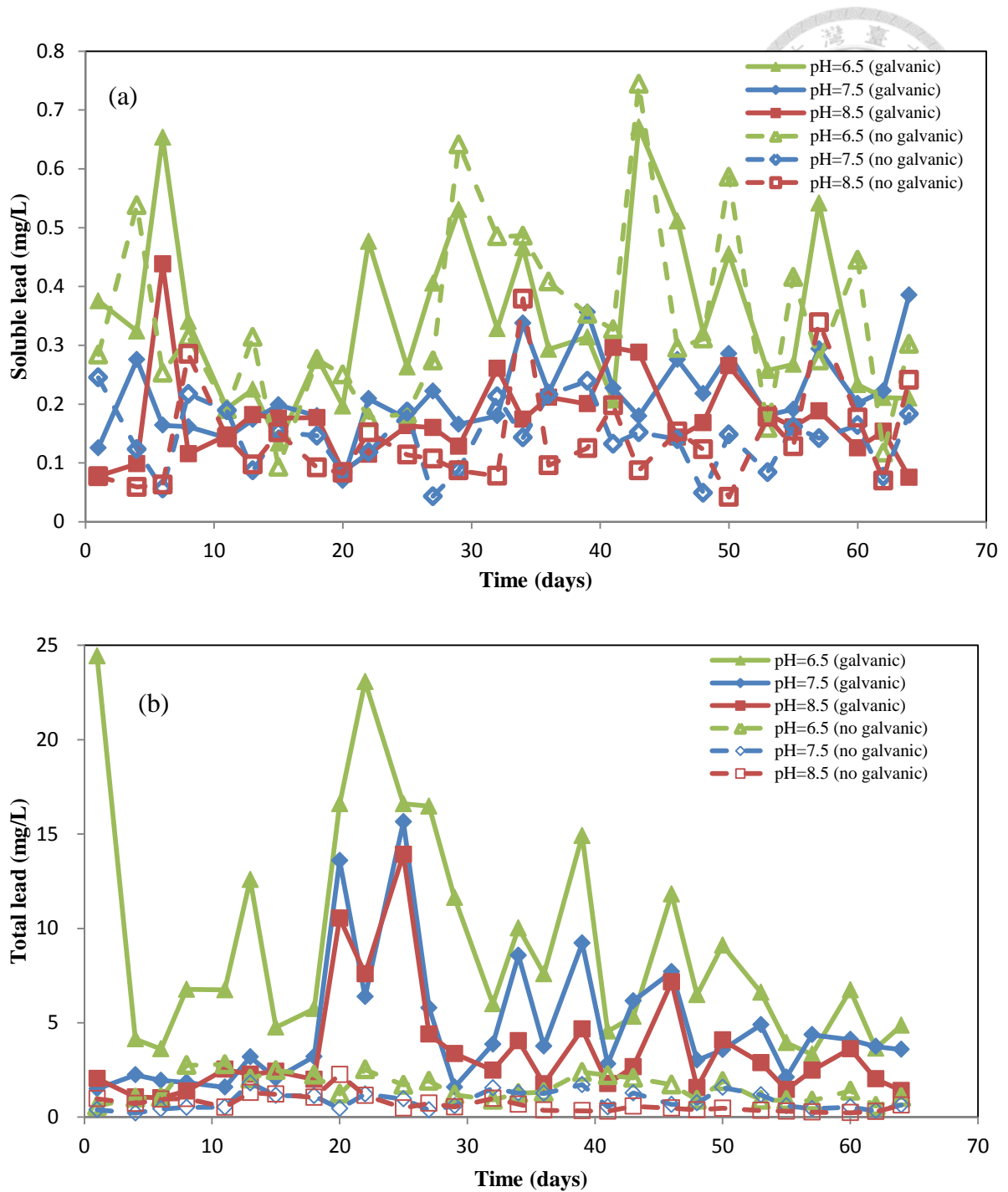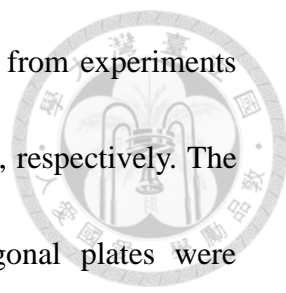
Figure 8 (a) Results of soluble lead concentration for experiments of determining the effects of pH on galvanic corrosion. (b) Results of total lead concentration for experiments of determining the effects of pH on galvanic corrosion.

The SEM and elemental mapping of the precipitates obtained from experiments conducted at pH 6.5, 7.5 and 8.5 are shown in Figure 9, 10 and 11, respectively. The SEM images showed that both rod shape particles and hexagonal plates were observed at pH 6.5 and 7.5, while only hexagonal plates were found at pH 8.5. The mapping data showed that both precipitates contained carbon, lead and oxygen. Based on the shapes of Pb(II) particles reported in the literature (Zhang and Lin, 2011) and the elemental mapping results, it can be concluded that the rod shape crystal is cerussite ($PbCO_3$) and the plate shape crystal is hydrocerussite ($Pb_3(CO_3)_2(OH)_2$). The formation of both cerussite and hydrocerussite at pH < 7.5 and the formation of only hydrocerussite at pH=8.5 are consistent with their thermodynamic stability under these pH values (Lytle and Schock, 2005).
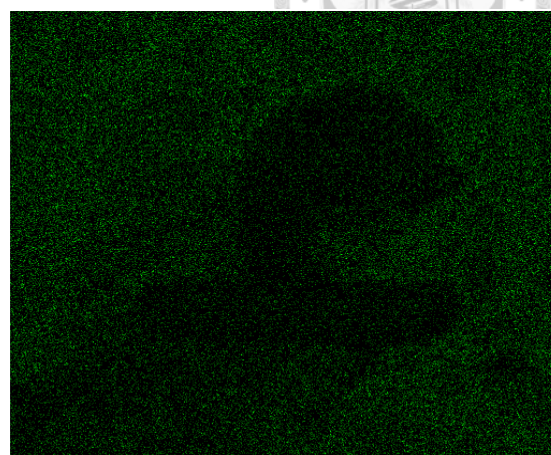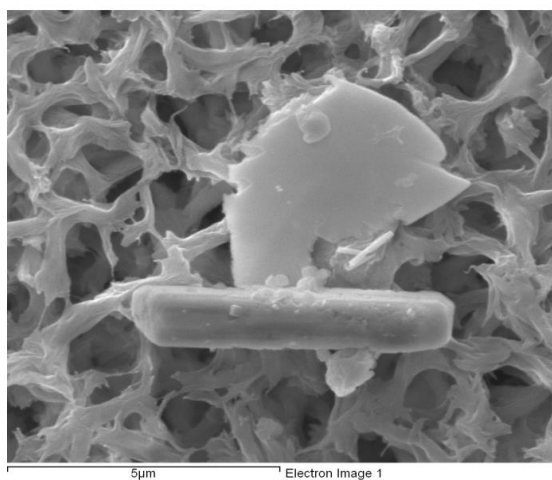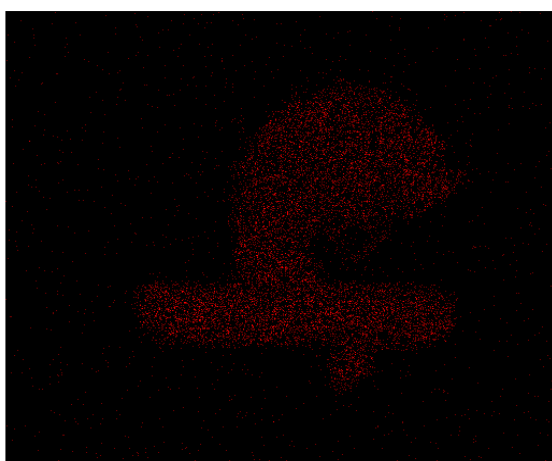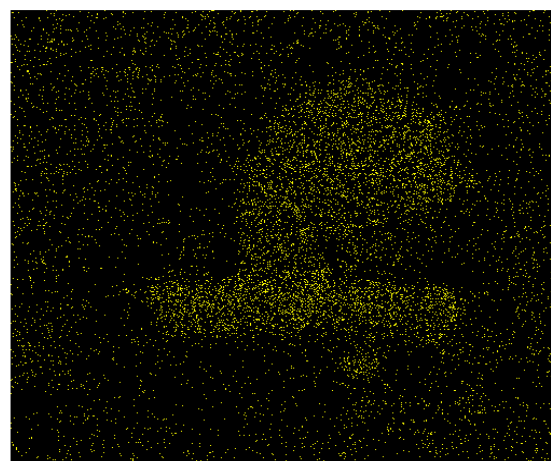
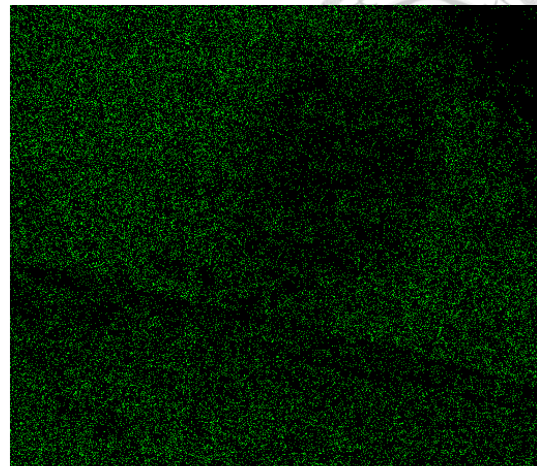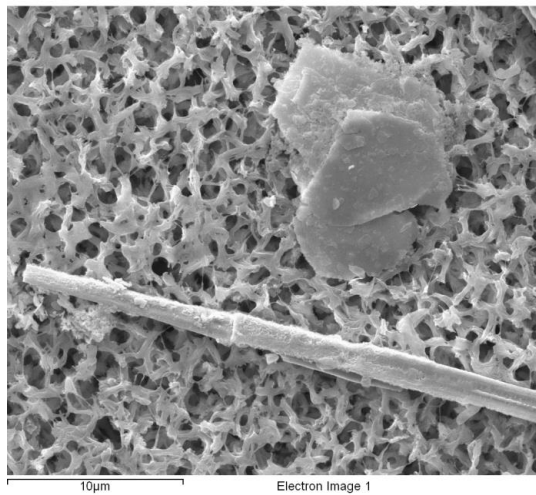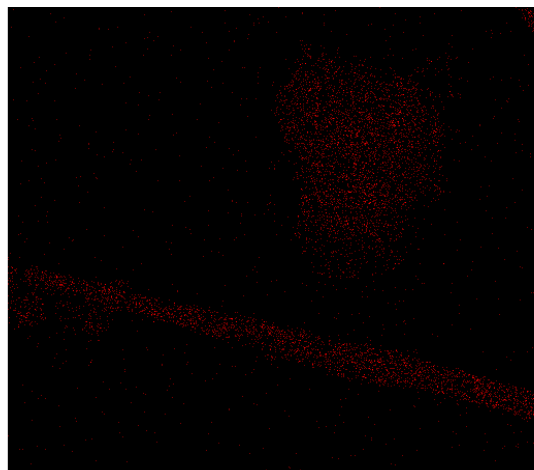Figure 9 SEM-EDX for particles collected from experiments conducted at pH=6.5, CSMR=0.5.

Figure 10 SEM-EDX for particles collected from experiments conducted at pH=7.5, CSMR=0.5.
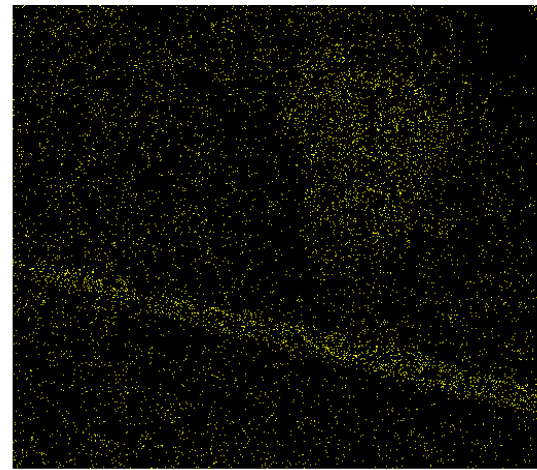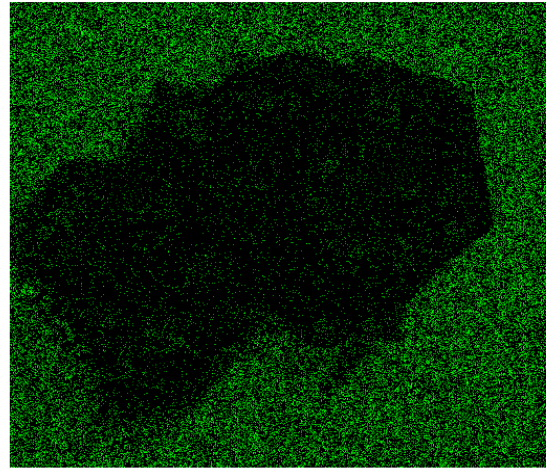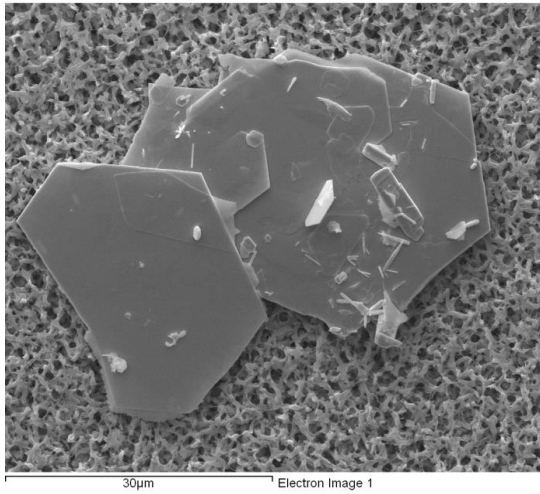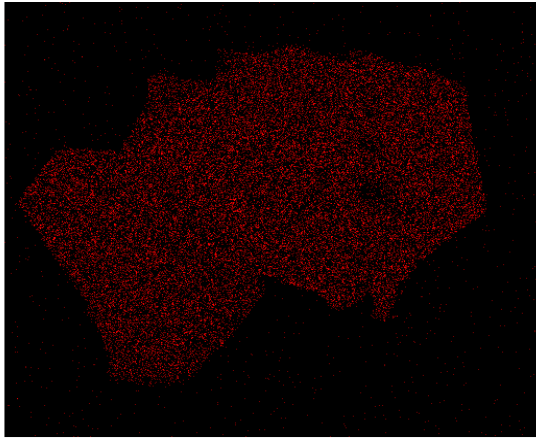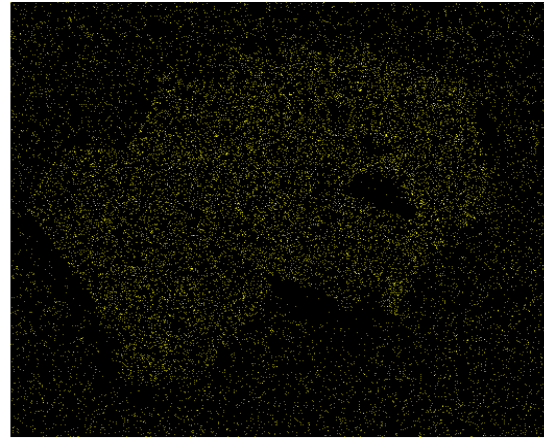
Figure 11 SEM-EDX for particles collected from experiments conducted at pH=8.5, CSMR=0.5.

4.4 Effects of varying chloride and sulfate mass concentration at a fixed CSMR

The release of soluble and total lead for the experiments conducted under a fixed CSMR of 2 but various chloride and sulfate mass concentrations are shown in Figure 12, in which CSMR=2 (Low) represents $[Cl^-]$=60 mg/L, $[SO_4^{2-}]$=30 mg/L, CSMR=2 (Medium) represents $[Cl^-]$=120 mg/L, $[SO_4^{2-}]$=60 mg/L and CSMR=2 (High) represents $[Cl^-]$=250 mg/L, $[SO_4^{2-}]$=125 mg/L.

In addition to forming precipitates, the enhanced release of Pb^{2+} caused by galvanic corrosion may form soluble complex such as $PbCl^+$ and $PbSO_4^0$. Lead precipitation can decrease soluble lead concentration while the formation of soluble lead complex may increase the soluble lead level. Therefore, it was expected that soluble lead concentration should increase with the increasing Cl^- and SO_4^{2-} concentration under a fixed CSMR. The obtained soluble lead results (Figure 12(a)), however, showed the opposite trend. Similarly, same trend was observed for the total lead concentration (Figure 12(b)). The possible explanation for the unexpected results is that when the concentration of Cl^- and SO_4^{2-} increased, excess Pb^{2+} was released initially to rapidly form massive precipitation attached on the pipe surfaces and blocked the sites for electron transfer, in turn, reduced the rate of galvanic corrosion. Future research can be devoted to resolving the mechanisms behind this observation.

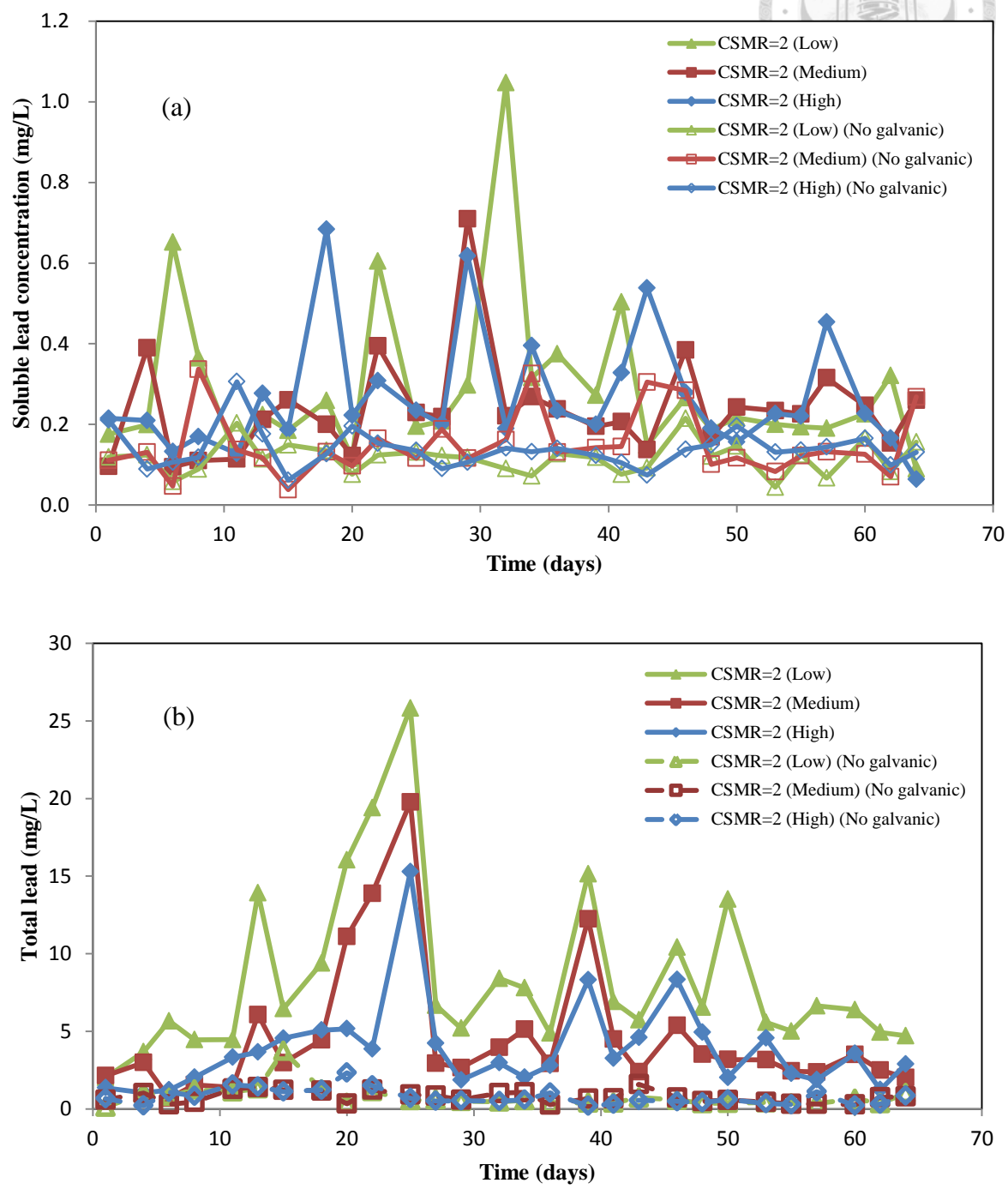Figure 12 (a) Soluble lead and (b) total lead concentration as a function of time for the experiments of varying the chloride and sulfate concentration under a fixed CSMR=2.

4.5 Effects of chloride and sulfate mass ratio (CSMR) and orthophosphate on galvanic corrosion.

The effects of CSMR and orthophosphate addition on the release of soluble lead and total lead are shown in Figure 13. The experiments were conducted for a total period of 18 weeks, in which the first 9 weeks allowed galvanic corrosion to proceed at different CSMR to observe the effects of varying CSMR on lead release, followed by a 5-week period of 1mg/L P orthophosphate addition to determine the effectiveness of using orthophosphate as a lead corrosion inhibitor, and finished with a 4-week period that the orthophosphate addition was stopped to evaluate the need of continuous addition of orthophosphate to suppress lead level.

The results from the first period indicated that both soluble and total lead release increased with the increasing CSMR (increased chloride concentration under a fixed sulfate concentration). This trend could be due to the higher formation of $PbCl^+$ complex when the chloride ion concentration increased, which in turn resulted in more precipitate formation. In addition, the high chloride concentration also increased the conductivity of the solution which could promote the rate of galvanic corrosion and increase the lead release.

The results obtained from the second period showed that both soluble and total lead concentrations were reduced, which could be due to the formation of

phosphate-based Pb precipitates. Results of SEM-EDX for the solids collected during orthophosphate addition are shown in Figure 14. The SEM image showed that there were hexagonal rod structures formed and the elemental mapping indicated that these structures contained P. The hexagonal rod crystal is likely to be chloropyromorphite due to the relative high chloride concentration employed in the experiments (Zhang et al., 1997). Based on the results, it can be concluded that orthophosphate can help to reduce the soluble and total lead release by forming precipitates with very low solubility.

The results obtained from the last period showed that both soluble and total lead concentrations began to rise after the orthophosphate addition was stopped. Therefore, a continuous addition of orthophosphate is required to suppress both soluble and total lead level if galvanic corrosion occurs
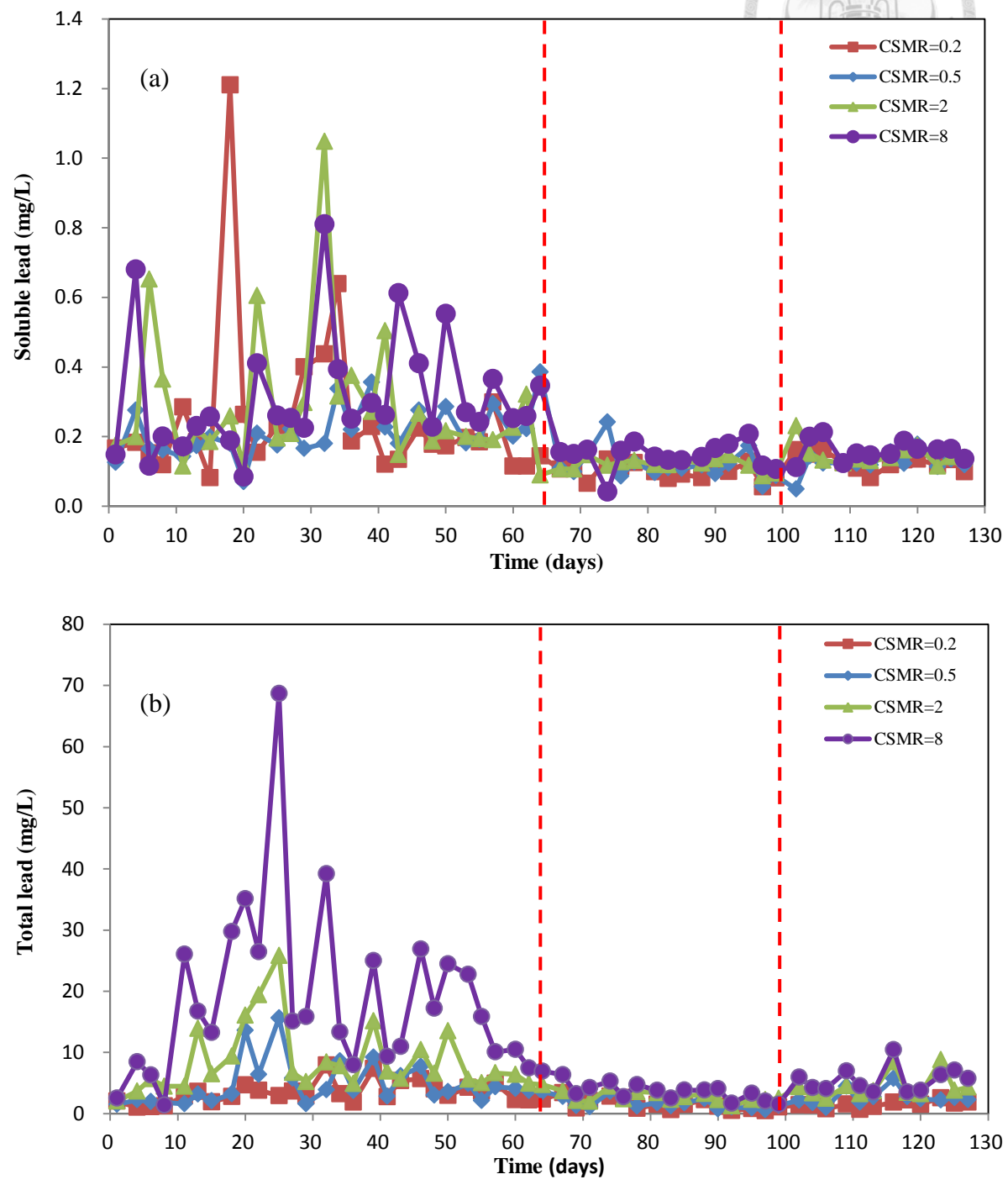
Figure 13 Results for experiments conducted for determining the effects of CSMR and orthophosphate on galvanic corrosion (a) Soluble lead (b) Total lead.
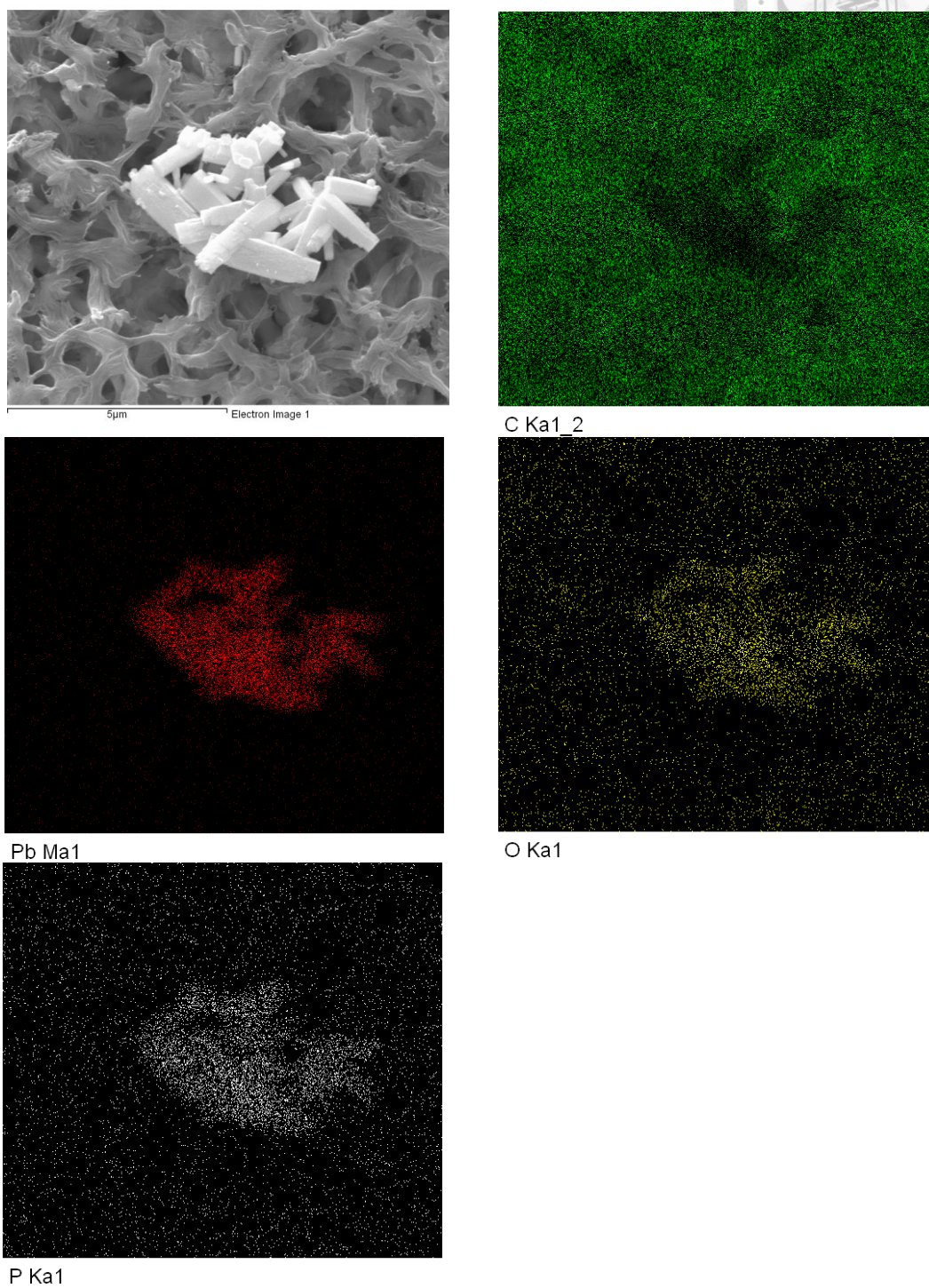
Figure 14 SEM-EDX for particles collected from experiments with orthophosphate addition (pH=7.5, CSMR=8).

Chapter 5 Conclusions and Recommendations

5.1 Conclusions

The influences of water chemistry on lead release resulting from galvanic corrosion of lead and stainless steel and the effectiveness of using orthophosphate as a corrosion inhibitor were investigated in this study. The conclusions obtained are summarized below:

1. Partial lead service line replacement with stainless steel materials could cause massive lead release due to galvanic corrosion and the lead release was mostly contributed by particulate lead.

2. The water was more corrosive under a lower pH, enhancing the release of both soluble and total lead in the galvanic corrosion.

3. A higher chloride and sulfate concentration under a fixed CSMR was found to unexpectedly decrease the lead release from galvanic corrosion and this phenomenon requires further investigation.

4. Tap water with a higher CSMR could be more corrosive possibly due to the formation $PbCl^+$ complex and a higher conductivity in the galvanic corrosion.

5. The addition of orthophosphate could reduce both soluble and total lead release by forming lead phosphate minerals. However, continuous addition of

orthophosphate is required to maintain a low lead level.

5.2 Recommendations for future study

Further studies that can provide more insights on lead release due to galvanic

corrosion are suggested as the followings:

1. Due to the unexpected results in the experiment of varying chloride and sulfate

    concentration under fixed CSMR=2, more studies are needed to resolve the

    mechanisms behind this observation.

2. The galvanic corrosion test between lead and stainless steel can be conducted

    under flow system in order to simulate the distribution system.

3. Lead and stainless steel could be separated with a nonconductive material and

    galvanically connected with an external copper wire to measure the galvanic

    current during the experiments.

4. Field studies will be needed to confirm the real situation of galvanic corrosion in

    the drinking water distribution system.

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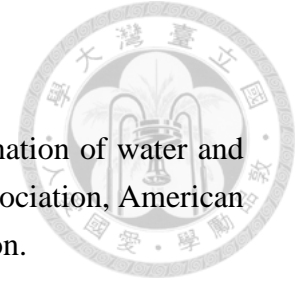
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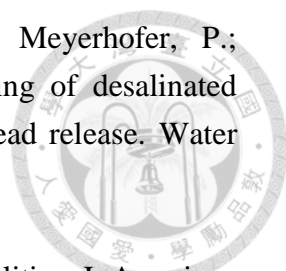
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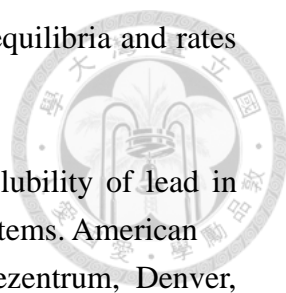
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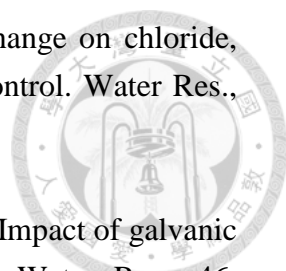
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